

The Optical Properties of the Giant Dielectric Response Material $\text{CaCu}_3\text{Ti}_4\text{O}_{12}$

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Beamline(s): U10A

Introduction: The cubic perovskite-like material $\text{CaCu}_3\text{Ti}_4\text{O}_{12}$ (group $Im\bar{3}$) exhibits a large dielectric response, the likes of which have not been seen, to our knowledge, in any existing material. This compound possesses a low-frequency dielectric constant of approximately 10^4 , which is only weakly varying in the temperature range 100-400 K. However, below 100 K there is an abrupt 100-fold reduction in the low-frequency dielectric constant¹. X-ray diffraction and thermoelectric data argue against an explanation in terms of ferroelectricity² (the collective ordering of local dipole moments).

Methods and Materials: A single crystal boule of $\text{CaCu}_3\text{Ti}_4\text{O}_{12}$ was grown using the traveling floating zone technique. A disc approximately 3 mm in diameter was cut from near the center of the boule and polished with progressive laps until a final polish with 0.1 micron diamond paste was applied. The final surface was optically flat and had a "bright" finish. The sample was then mounted on the top of an optically-black cone³. The reflectance of $\text{CaCu}_3\text{Ti}_4\text{O}_{12}$ was measured over a wide frequency range (from approximately 20 cm^{-1} to 8000 cm^{-1} (3 meV to 1 eV, where 1 eV is 8065 cm^{-1}) with the Bruker IFS66v/S spectrometer at U10A using an *in situ* overcoating technique that has been described in detail elsewhere³.

Results: The reflectance of $\text{CaCu}_3\text{Ti}_4\text{O}_{12}$ shown in **Figure 1** in the region of interest. The reflectance above 800 cm^{-1} shows little frequency dependence, while at low frequency the reflectance is increasing with decreasing temperature. The optical conductivity has been calculated from a Kramers-Kronig analysis of the reflectance, and is shown in **Figure 2** for the low-frequency region (the inset shows the conductivity at room temperature over a wider range, where a total of 10 infrared active vibrations are observed). The quartet of low-frequency modes shows some unusual behavior. Not only do they soften instead of hardening with decreasing temperature (unlike the remaining high-frequency modes), but they also appear to develop shoulder-like features indicative of splitting. The characteristic temperature at which these changes occur is about 100 K, which is also where the low-frequency dielectric constant under-goes a rapid change. The oscillator strengths also appear to increase in an anomalous manner at low temperature.

Conclusions: Infrared spectroscopy has shown clear changes near the temperature associated with large changes in the dielectric constant, which will play an important role in understanding the changes that are occurring in this material.

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References: ¹ M.A. Subramanian, D. Li, N. Duan, B.A. Reisner, A.W. Sleight, J. Solid State Chem. **151**, 323 (2000); ² A.P. Ramirez, M.A. Subramanian, M. Gardel, G. Blumberg, D. Li, T. Vogt, and S.M. Shapiro, Solid. State Commun. **115**, 217 (2000); ³ C.C. Homes, M. Reedyk, D.A. Crandles, and T. Timusk, Appl. Optics **32**, 2976 (1993).

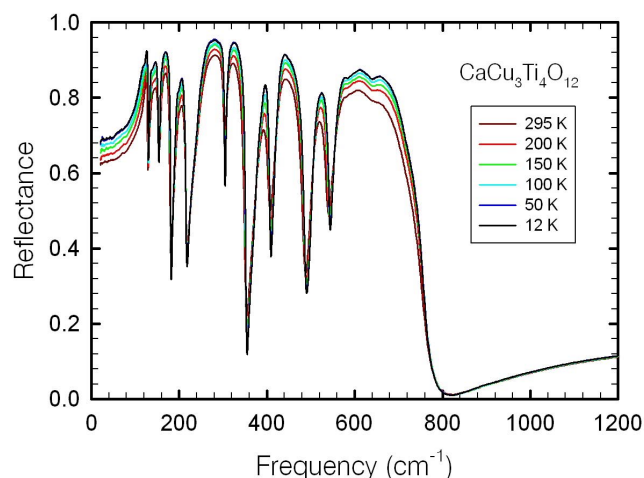


Figure 1. The reflectance of $\text{CaCu}_3\text{Ti}_4\text{O}_{12}$ in the far and mid-infrared region, at several different temperatures.

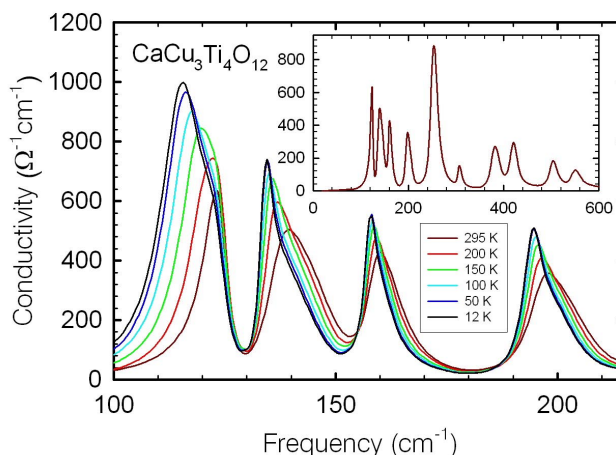


Figure 2. The optical conductivity of $\text{CaCu}_3\text{Ti}_4\text{O}_{12}$ determined from a Kramers-Kronig analysis of the reflectance shown in Figure 1.